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Nickel isotope fractionation during laterite Ni ore smelting and refining: Implications for tracing the sources of Ni in smelter-affected soils

G. Ratié^{a, b, *}, C. Quantin^a, D. Jouvin^a, D. Calmels^a, V. Ettler^c, Y. Sivry^d, L. Cruz Vieira^b, E. Ponzevera^e, J. Garnier^b

^a UMR 8148 GEOPS, Univ. Paris Sud – CNRS-Université Paris-Saclay, 91405 Cedex, France

^b UnB, IG/GMP-ICC Centro, Campus Universitario Darcy Ribeiro, 70910-970, Brasília-DF, Brazil/Laboratoire Mixte International, LMI OCE « Observatoire des Changements Environnementaux », Institut de Recherche pour le Développement/University of Brasília, Campus Darcy Ribeiro, Brasília, Brazil

^c Institute of Geochemistry, Mineralogy and Mineral Resources, Charles University in Prague, Albertov 6, 12843 Prague 2, Czech Republic

^d Institut de Physique du Globe de Paris, Sorbonne Paris Cité, Univ. Paris Diderot, UMR 7154 CNRS, F-75005 Paris, France

^e IFREMER, Centre de Brest, Unité Géosciences Marines, 29280, Plouzané, France

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ABSTRACT

Nickel isotope ratios were measured in ores, fly ash, slags and FeNi samples from two metallurgical plants located in the Goiás State, Brazil (Barro Alto, Niquelândia). This allowed investigating the mass-dependent fractionation of Ni isotopes during the Ni-laterite ore smelting and refining. Feeding material exhibits a large range of $\delta^{60}\text{Ni}$ values (from $0.02 \pm 0.10\%$ to $0.20 \pm 0.05\%$, $n = 7$), explained by the diversity of Ni-bearing phases, and the average of $\delta^{60}\text{Ni}_{\text{feeding materials}}$ was found equal to $0.08 \pm 0.08\%$ (2SD, $n = 7$). Both $\delta^{60}\text{Ni}$ values of fly ash ($\delta^{60}\text{Ni} = 0.07 \pm 0.07\%$, $n = 10$) and final FeNi produced ($0.05 \pm 0.02\%$, $n = 2$) were not significantly different from the feeding materials ones. These values are consistent with the very high production yield of the factories. However, smelting slags present the heaviest $\delta^{60}\text{Ni}$ values of all the smelter samples, with $\delta^{60}\text{Ni}$ ranging from $0.11 \pm 0.05\%$ to $0.27 \pm 0.05\%$ ($n = 8$). Soils were also collected near and far from the Niquelândia metallurgical plant, to evaluate the potential of Ni isotopes for tracing the natural vs anthropogenic Ni in soils. The Ni isotopic composition of the non-impacted topsoils developed on ultramafic rocks ranges from $-0.26 \pm 0.09\%$ to $-0.04 \pm 0.05\%$ ($n = 20$). On the contrary, the Ni isotopic composition of the non-ultramafic topsoils, collected close to the plant, exhibit a large variation of $\delta^{60}\text{Ni}$, ranging from $-0.19 \pm 0.13\%$ up to $0.10 \pm 0.05\%$ ($n = 4$). This slight but significant enrichment in heavy isotopes highlights the potential impact of smelting activity in the surrounding area, as well as the potential of Ni isotopes for discerning anthropogenic samples (heavier $\delta^{60}\text{Ni}$ values) from natural ones (lighter $\delta^{60}\text{Ni}$ values). However, given the global range of published $\delta^{60}\text{Ni}$ values (from -1.03 to 2.5%) and more particularly those associated to natural weathering of ultramafic rocks (from -0.61 to 0.32%), the use of Ni isotopes for tracing environmental contamination from smelters will remain challenging.

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1. Introduction

Nickel is an important metal in modern infrastructure and technology, with major uses in the production of stainless steel (60% of the global primary Ni consumption, Nickel Institute, 2013)

and alloys and other application such as electroplating or production of rechargeable batteries (Mudd, 2010). It is therefore of major economic importance. The worldwide increasing demand of metals for economic purpose induces intense mining that may lead to anthropogenic forcing on the environment. The world nickel production increased from 1,400,000 t to 2,400,000 t over the last ten years (USGS, 2015). For instance, the Chinese stainless steel industry was the main consumer of primary Ni in 2011 (521,000 t of primary Ni), increasing its Ni demand by 22% in one year (USGS, 2011). Economic Ni resources are found either in magmatic

* Corresponding author. UMR 8148 GEOPS, Université Paris Sud – CNRS, 91405 Cedex, France.

E-mail address: gildas.ratie@gmail.com (G. Ratié).

sulfide deposits, where the principal ore mineral is pentlandite ((FeNi)₉S₈) (Naldrett et al., 1979; Hoatson et al., 2006), or in laterite-type ores deposits where the main ore minerals are nickeliforous limonite and garnierite (hydrated Mg–Ni phyllosilicates, e.g. (Mg,Ni)₃Si₂O₅(OH)₄) (Dalvi et al., 2004; Butt and Cluzel, 2013). Currently, about 1/3 of the primary Ni is produced by smelting of lateritic ores, which represent 60–70% of the terrestrial reserves, and is used at 90% for ferronickel (FeNi) and 10% for sulfide matte (Crundwell et al., 2011). To meet future demand for Ni, laterite ores are increasingly mined.

The feeding material of laterite smelting corresponds to a mixture of moist saprolitic and lateritic ores derived from the weathering of ultramafic (UM) rock under tropical conditions. Ores developed in such UM rocks contain 0.4 to 3 wt.% Ni (Dalvi et al., 2004; Butt and Cluzel, 2013). In saprolitic ores, Ni is commonly found associated with primary and secondary clay minerals such as serpentine, talc, chlorite and smectite (Colin et al., 1985; Manceau and Calas, 1985; Gaudin et al., 2004; Gleeson et al., 2004). In some saprolite veins, talc-like and serpentine-like minerals, also known as “garnierite”, may contain up to 30 wt.% Ni (Wells et al., 2009; Villanova-de-Benavent et al., 2014). Sepiolite may also contain significant Ni amounts (Gleeson et al., 2004; Villanova-de-Benavent et al., 2014; Ratié et al., 2015). In the lateritic ores, Fe-oxides are the main Ni-bearing minerals (Manceau et al., 2000; Quantin et al., 2002; Dublet et al., 2012 and 2015) and can residually concentrate Ni up to 1 wt. %. Moreover, in the transition laterite, Mn-oxides can also be significant Ni-scavengers (Fan and Gerson, 2011). Due to the high Fe content of the saprolitic and limonitic ores mixture (up to 40 wt.% Fe₂O₃), the smelting product is FeNi, for which typical composition is 20–40% Ni and 80–60% Fe, after refining (Crundwell et al., 2011).

The FeNi production from laterite smelting follows four main steps: drying of the feeding material in rotating kiln, calcination

with coal, oil or other organic products, reduction in electric furnace and finally, refining of the molten FeNi in an electric furnace (Crundwell et al., 2011). These smelting steps also produce a huge amount of by-products (fly ash, smelting slags and refining slags), which may contain significant amounts of Ni and other trace metals (see Table 1). Waste materials are either stored in settling ponds (fly ash) or dumped (slags) or partially reprocessed (fly ash). Such disposal sites might have environmental consequences in the vicinity of the smelters, due to the release of Ni and other metals by leaching and erosion during long-term storage (Ettler et al., 2005). Moreover, the widespread use of Ni in industry is responsible for significant anthropogenic discharge in the environment (Romic and Romic, 2003; Buzier et al., 2011).

The isotopic composition of metals has been shown to constitute an efficient tool to trace anthropogenic contamination in the environment (e.g. Cloquet et al., 2006; Weiss et al., 2008; Chen et al., 2008; Mattioli et al., 2009; Stetson et al., 2009). Isotope fractionation of Zn, Cu, Cd and Hg has been observed during roasting, smelting and refining processes (Mattioli et al., 2006; Sivry et al., 2008; Sonke et al., 2008; Shiel et al., 2010; Gray et al., 2013). For instance, the Zn remaining in metallurgical tailings is significantly fractionated relative to Zn ore, allowing to trace anthropogenic Zn contamination in the Lot watershed, France (Sivry et al., 2008). Zinc isotopes have also been used to evidence the imprint of smelting activities on the surrounding environment through atmospheric deposition of smelting residues (Sonke et al., 2008).

This article aims at determining nickel isotopic fractionation associated with smelting processes of Ni laterite ore in two operating plants located in the Goiás State, Brazil. Our study reports the first Ni isotope data on feeding materials, products and waste materials involved in the different steps of a metallurgical process. The second objective of the paper is to evaluate the potential use of

Table 1
Total contents of elements and Ni isotopic composition ($\delta^{60}\text{Ni}$, $\delta^{62}\text{Ni}$) of mixed Ores (Ore), Fly ash (F), Smelting Slags (SS, ^a old, ^b recent), White Refining Slags (WRS), Black Refining Slags (BRS), FeNi.

Sample Name	Ca	Mg	Fe	Mn	Ni	$\delta^{60}\text{Ni}$	2SD	$\delta^{62}\text{Ni}$	2SD	
						g/kg				‰
Niquelândia	Ore1	2.54	92.3	165	2.69	16.9	0.03	0.05	0.06	0.10
	Ore2	1.79	97.3	148	2.42	23.2	0.03	0.05	0.06	0.09
	Ore3	3.89	81.4	135	2.56	20.1	0.20	0.05	0.40	0.10
	Ore4	2.70	103	151	2.89	17.6	0.05	0.05	0.06	0.12
	F1	3.33	88.7	157	3.23	22.7	0.01	0.05	0.02	0.10
	F2	2.85	90.6	193	3.24	27.0	0.03	0.06	0.06	0.12
	F3	3.48	110	200	3.53	27.6	0.01	0.05	0.03	0.10
	F4	3.30	73.5	209	2.9	24.6	0.06	0.08	0.11	0.15
	F5	3.41	70.5	245	3.99	23.6	0.03	0.05	0.05	0.10
	F6	1.90	92.4	178	5.13	22.9	0.15	0.06	0.28	0.14
	F7	2.60	17.3	365	4.63	8.30	0.01	0.05	0.02	0.10
	F8	5.10	20.9	311	3.48	7.80	0.05	0.05	0.10	0.10
	F9	2.77	93.0	285	3.84	18.9	0.20	0.05	0.38	0.10
	SS1 ^a	12.83	55.1	131	2.39	1.81	0.11	0.05	0.21	0.10
	SS2 ^a	5.71	188	117	3.17	1.90	0.16	0.05	0.31	0.10
	SS3 ^b	2.68	161	111	2.86	1.17	0.20	0.05	0.39	0.10
	SS4 ^b	3.97	179	143	3.44	1.59	0.27	0.05	0.52	0.14
	SS5 ^a	2.20	155	122	2.89	0.98	0.17	0.11	0.31	0.25
	FeNi1	1.86	<LD	661	<LD	338	0.04	0.05	0.08	0.10
Barro Alto	Ore5	1.93	95.9	118	2.62	18.5	0.02	0.10	0.05	0.20
	Ore6	3.45	110	164	3.15	18.1	0.17	0.13	0.33	0.26
	Ore7	1.60	71.4	178	3.02	22.2	0.04	0.06	0.07	0.13
	F10	4.88	123	265	3.86	30.9	0.10	0.12	0.20	0.23
	SS6 ^b	4.19	187	68.8	2.95	1.64	0.18	0.05	0.35	0.10
	SS7 ^b	2.90	162	106	2.60	1.14	0.22	0.05	0.44	0.10
	SS8 ^b	3.42	153	124	2.89	2.03	0.14	0.05	0.26	0.10
	WRS	69.5	143	71.2	1.97	2.28	0.14	0.06	0.27	0.13
	BRS	149	102	179	1.59	6.28	0.03	0.06	0.06	0.12
	FeNi2	0.30	<LD	689	<LD	311	0.07	0.06	0.13	0.13

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